

# Comment on “Transition from Bose glass to a condensate of triplons in $Tl_{1-x}K_xCuCl_3$

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We argue that the interpretation of the calorimetric data for disordered quantum antiferromagnets  $Tl_{1-x}K_xCuCl_3$  in terms of Bose Glass physics by F. Yamada *et al.* in [Phys. Rev. B **83**, 020409(R) (2011)] is not unambiguous. A consistent analysis shows no difference in the crossover critical index for the disorder-free  $TlCuCl_3$  and its disordered derivatives. Furthermore, we question the very existence of a proper field-induced thermodynamic phase transition in  $Tl_{1-x}K_xCuCl_3$ .

The main claim of the paper by F. Yamada *et al.* is that K-substitution in  $Tl_{1-x}K_xCuCl_3$  changes the field-induced Mott Insulator (MI) to BEC (Bose-Einstein Condensate) transition in the pure system to a Bose Glass (BG) to BEC transition in the chemically disordered compound. This conclusion is primarily drawn from a comparison of the crossover exponents  $\phi$ . We question the validity of such an analysis.

First, the data range is too small: at best it covers half a decade in temperature, and as little as a quarter of a decade for some cases. Second, the error of determining the peak position in field scans is actually quite significant. The scattering of the raw data that appears in the top three curves on Fig. 1b gives an estimate of the noise in specific heat measurements: about  $10^{-3}$  J/mol K. The method used for locating the maximum from noisy data is not described in the paper. However, given the widths of the peaks, the noise must translate into an error of determining the maximum position of *at least*  $\pm 0.1$  T at high temperature (as admitted in the paper) and as much as  $\pm 0.2$  T at low temperatures. To illustrate the impact of these factors we have re-analyzed the data digitized from Fig. 1c assuming an arbitrarily fixed exponent  $\phi = 1.0$ , and a very small measurement error of 0.05 T in the entire range. The fits yield almost perfect agreement for *all three compositions*:  $\chi^2 = 0.25$ ,  $\chi^2 = 0.38$  and  $\chi^2 = 0.22$  for  $x = 0$ ,  $x = 0.22$  and  $x = 0.36$ , respectively, and a confidence of determination  $R^2 > 0.985$  in all cases.

The rather small least-squares errors quoted for  $\phi$  are actually a poor measure of the confidence interval. In fact,  $\phi$  has very high covariance with another fit parameter, namely the critical field. The latter is not measured independently. We found that the covariance becomes particularly large when the fitting range is restricted to low temperatures.

A meaningful comparison between the three compositions requires that the same data range or the same decreasing fitting window procedure is used in each case in a *consistent* analysis. The authors obtain  $\phi = 1.53$  in the pure compound by including all data up to 2 K. Yet, the BG-to-BEC value  $\phi = 0.58$  in the  $x = 0.36$  material is only found when the fitting range is restricted to be-

low 1 K. That such a free choice of fitting window can be misleading, is illustrated by the following. Re-fitting digitized author’s data for  $T > 0.6$  K (with only four low-temperature points excluded) yields MI-to-BEC-like exponents for *both*  $x = 0$  and  $x = 0.36$ :  $\phi = 1.74(0.2)$  and  $\phi = 1.39(17)$ , respectively. Thus, all conclusions of the paper fully rely on *just these 4 data points*. The latter to correspond to the poorest-defined maxima.

A related issue concerns the  $x = 0.22$  data for which the authors altogether avoid discussing the decreasing fitting window analysis. Such an analysis of digitized data shows that  $\phi$  remains close to unity and exhibits no systematic decrease with decreasing  $T_{max}$ . For  $T_{max} = 1$  K we get  $\phi = 1.02(0.3)$ .

The final issue concerns the broad specific heat peaks in the raw  $C(H)$  data. A sharp lambda-anomaly that is a hallmark of a continuous phase transition is entirely absent. It is conceivable that such peak broadening is instrumental. Since the instrumental function is not known, the transition point can not be unambiguously associated with the measured maximum. The actual relation will depend on the temperature dependence of the underlying specific heat divergence, the measurement method and other factors. The peaks being broad as they are, an 0.1 T mismatch at low temperatures can not be excluded. The result would be a temperature-dependent systematic error that directly affects  $\phi$ .

Alternatively, the specific heat of the samples actually *does not diverge*. This implies that there is no continuous thermodynamic phase transition, and that the correlation length remains finite. Under these circumstances, any discussion of “criticality” or critical indexes loses its significance. The absence of a true phase transition may itself be a result of chemical disorder or due to a staggered g-tensor and Dzyaloshinskii-Moriya interactions.<sup>1</sup>

Considering what is said above, it is not surprising that “the BEC phase is not reached even at the lowest temperature” in ESR experiments. What is surprising, is that it is at these very temperatures (0.55 K) the authors claim to see evidence of the BEC phase in bulk measurements.

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<sup>1</sup> J. Sirker, A. Weiße and O. P. Sushkov, Europhys. Lett. **68**, 275(2004).